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NUCLEAR POLARIZATION OF A ^3He GAS BY LASER OPTICAL PUMPING

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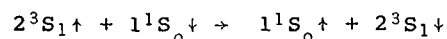
Résumé.- On donne une brève introduction sur le pompage optique comme méthode de polarisation nucléaire d'un gaz de ^3He . On devrait pouvoir produire de fortes polarisations en remplaçant les lampes à décharge par des lasers accordables comme sources de pompage. On discute brièvement les problèmes posés par la source laser. On donne les résultats de deux tentatives expérimentales : pompage synchrone en picoseconde d'un laser à colorants infra-rouge, et pompage en tandem d'un laser à centres colorés du type F_2^+ nouvellement découverts.

Abstract.- A brief introduction to the optical pumping technique as a method for nuclearly polarizing a ^3He gas is given. Large polarizations should be obtainable by replacing the discharge lamps by tunable lasers as pumping sources. The characteristics of a suitable laser source are briefly discussed. Results are given for two experimental attempts : picosecond synchronous pumping of I R dye lasers and tandem pumping of newly discovered F_2^+ like colored center lasers.

The physical properties of a system of ^3He atoms with all nuclear spins parallel (fully polarized ^3He , or $^3\text{He}^+$) can be significantly different from those of non-polarized ^3He /1/. The origin of these differences arises from the Pauli exclusion principle. For instance, important modifications of the transport properties of the gas (viscosity, heat conduction) at low temperatures are expected, for a 100% nuclear polarization. At lower nuclear polarization, the modifications are only second order effects which are more difficult to observe experimentally. This is the reason why an effective method to obtain high polarization in ^3He is needed, and we have started a program to improve the efficiency of optical pumping by using a laser instead of a discharge lamp as a light source. After briefly recalling the principle of optical pumping in a ^3He gas, we report on our preliminary results obtained with two different kinds of lasers.

The possibility of optically creating a nuclear orientation of an ensemble of ^3He atoms in the ground state was first demonstrated by Colegrove et al /2/. The method is a non direct optical pumping process. As it is not possible, for experimental and theoretical reasons, to use the resonance line $1^1S_0 - 2^1P$ for pumping, the idea

is to excite a number of atoms into the 2^3S_1 metastable state by a weak discharge in the cell (see fig. 1), and to optically pump these atoms with a circularly polarized light beam at $\lambda = 1\ 083\ \text{nm}$ ($2^3S_1 - 2^3P$ transition, see fig. 1). The absorption of a σ^+ photon by the metastable atom, followed by spontaneous emission, results in a transfer of angular momentum from the photon to the atom. The metastable atom is thus left with an orientation of its electronic angular momentum, which is coupled to the nuclear spin through hyperfine interaction. It has been demonstrated /2/ that this process also induces a nuclear polarization of the ^3He atoms in the ground state. The reason is the occurrence of an effective transfer of orientation from the metastable to the ground state atoms, under the effect of the so-called metastability exchange collisions :



During such a collision, the nuclei of the two colliding particles exchange their electronic clouds ; the collision time is rather short and the nuclear spins are practically unaffected by the collision, so that the nuclear orientation is completely transferred from the 2^3S_1 metastable state to the 1^1S_0 ground state. The time

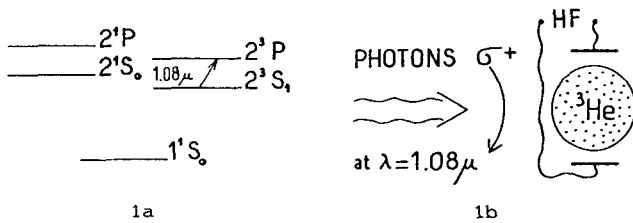


Fig. 1 : a) Level scheme of the first excited states of the helium atom. The 2^1S_0 and 2^3S_1 states are both metastable and are at about 20 eV above the 1^1S_0 ground state.

b) Principle of the optical pumping experiment in ^3He : a H.F. discharge in the ^3He cell carries a number of atoms into the 2^3S_1 metastable state. These metastable atoms are optically pumped by the 1.083μ line ($2^3P - 2^3S_1$ transition).

constant T_p for creation of the orientation inside the large reservoir of ^3He ground state atoms ($\sim 10^{17}$ atoms per c c) is a function of the collisional exchange time with the metastable atoms ($\sim 10^{10}$ atoms per c c) and of various relaxation processes in both levels. A convenient way to monitor the ground state nuclear orientation is to measure the circular polarization of some visible lines emitted by the cell /3/ : as a matter of fact, when nucleary oriented, ^3He atoms are excited from the ground state to any upper state by collisions in the discharge, their nuclear spins are unaffected, so that these excited atoms have a nuclear orientation too ; hyperfine interactions partially convert such a nuclear orientation into an electronic one. Subsequently, when these excited atoms decay through spontaneous emission, they emit circularly polarized photons. The rate of circular polarization of the emitted lines is thus proportional to the nuclear orientation of the ^3He ground state atoms and can be easily experimentally detected.

At 300 K, with conventional discharge lamps for pumping, T_p is typically of the order of a few minutes, and the ^3He nuclear polarizations obtained are usually $P \sim 5 - 20\%$. The limiting factor for obtaining large polarization in such experiments is the number of infrared pumping photons available per second. This is why we decided to replace the conventional discharge lamps by a tunable infrared laser at $\lambda = 1.083 \mu$, the estimated required power being of the order a few 10 mW within a spectral width

of 1 GHz. C.W. emission was preferred to pulsed emission with the same average power, in order to avoid stimulated emission from the 2^3P state, which would just cause the atoms to oscillate between the 2^3P and 2^3S_1 states, resulting in no additional polarization gain.

Our first attempt to obtain a C.W. tunable laser at 1.083μ was to extend the bandwidth of the existing IR dye lasers. However, with real C.W. operation, we did not succeed in obtaining wavelength longer than 1.04μ . We thus turned to synchronous pumping of the same IR dyes, by mode-locking the Kr^+ laser line at 752 nm and extending the dye laser cavity to the same length as the Kr^+ laser (synchronous pumping). In this way, the dye laser emitted picosecond pulses with a very high repetition rate (~ 100 MHz), and the oscillation threshold was substantially reduced (in average power), as compared to the C.W. case. Consequently, the tuning range of these IR dyes was significantly extended towards the infrared /4/.

We thus obtained a tunable dye laser at 1.083μ in picosecond operation with an infrared dye, IR 143, which unfortunately happens to be very unstable. We obtained an average power of 5 to 10 mWatt, inside a bandwidth of the order of 50 GHz, but only during a few hours due to the decay of the dye solution. Nevertheless we decided to try optical pumping, and ^3He nuclear polarizations up to 10% were obtained. To improve these results, we believe that better and more stable infrared dyes are required.

Very recently, L.F. Mollenauer demonstrated that a new kind of colored center lasers would work in C.W. operation at 1.083μ /5/. The emitting medium is a NaF crystal in which F_2^+ like centers are created by appropriate electronic bombardment. These centers are room-temperature stable. They have an absorption band centered at 870 nm and a broad emission band giving rise, when properly excited, to a laser emission ranging between 0.99 and 1.22μ /5/. With the help of L.F. Mollenauer, we used several such

crystals to obtain laser oscillation at 1.083μ through tandem pumping, as shown in figure 2 : a Kr^+ laser delivers a power of 3 Watts at 752 nm ; it excites a dye laser (HITC) which converts this wavelength into $\lambda = 870 \text{ nm}$, with about 20% efficiency ; this wavelength is well suited for the excitation of the colored centers. The crystal is maintained at liquid nitrogen temperature to avoid destruction of the centers under radiation. In a very preliminary experiment,

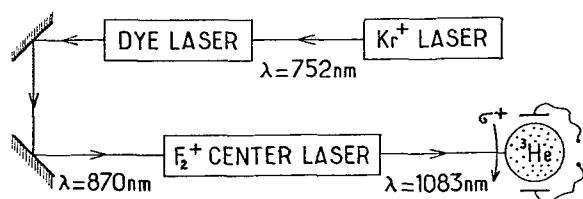


Fig. 2 : Diagram showing the experimental set up for optical pumping of a ^3He gas with a colored center laser : the 752 nm line of a Kr^+ laser is converted into $\lambda = 870 \text{ nm}$ by a H.I.T.C. dye laser, which in turn pumps a F_2^+ like colored center laser emitting tunable light a $\lambda = 1.084 \mu$.

a C.W. power of 30 mWatts, tunable over the 1.083μ region, was obtained. Figure 3 shows a ^3He nuclear polarization build-up signal recorded with such a laser and with the optical detection technique described above : at the middle of the recording, the polarization was suddenly destroyed by applying a NMR field (see fig. 3). Polarizations up to 30% have been observed, but higher values are hoped for the future. The colored center laser can be improved in several ways ; in particular, a single mode operation can be obtained by adding selective elements in the cavity. A systematic study of the optical pumping efficiency, when sweeping the unique laser mode over the full fine and hyperfine structure of the atomic line, is in progress. Improvement of the colored center laser efficiency is also expected. A more efficient excitation of this laser could be obtained by replacing the IR dye laser of the tandem pumping (see fig. 2) by another colored center laser /5/, resulting in more power at $\lambda = 870 \text{ nm}$. In all respects the signal recorded in figure 3 should thus be regarded as very preliminary.

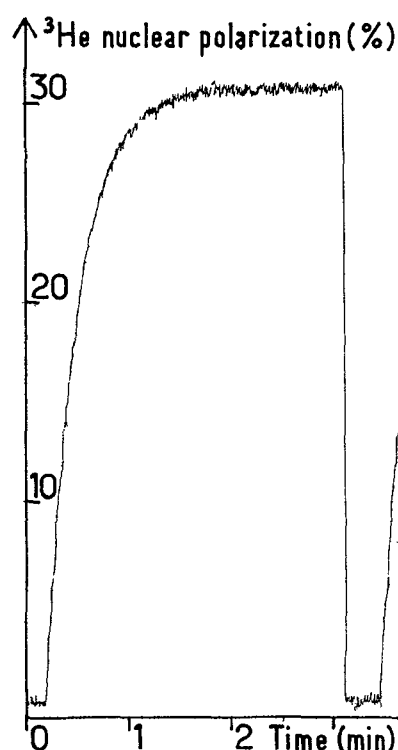


Fig. 3 : Nuclear polarization of a ^3He gas ($p = 0.3$ torr at 300 K) optically oriented by a colored center laser. One observes a build-up of the polarization with time, and its sudden destruction when a NMR field is applied.

When large nuclear polarizations are obtained by laser optical pumping, many interesting experiments will become possible. For example, the polarization of liquid ^3He by atom exchange with a polarized ^3He vapour, already observed long ago by M.C. Adams /6/, could be obtained in much better experimental conditions. Another improvement of this experiment should result from the use of cryogenic coatings (solid coatings of H_2 , Ne or liquid ^4He which minimize the nuclear relaxation of the gas on the walls of the container at low temperatures /7//8/). The change of the saturating vapour pressure, discussed in /1/, should then be observable. Another application of laser optical pumping would be to nuclearly polarize liquid ^3He solutions in ^4He , using the method developed by M.A. Taber /9/.

It is a great pleasure for us to thank L.F. Mollenauer whose help was quite decisive for our experiments.

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